Fluctuating Topological Defects in 2D Liquids: Heterogeneous Motion and Noise

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We measure the defect density as a function of time at different temperatures in simulations of a two dimensional system of interacting particles. Just above the solid to liquid transition temperature, the power spectrum of the defect fluctuations shows a $1/f$ signature, which crosses over to a white noise signature at higher temperatures. When $1/f$ noise is present, the 5-7 defects predominately form string like structures, and the particle trajectories show a 1D correlated motion that follows the defect strings. At higher temperatures this heterogeneous motion is lost. We demonstrate this heterogeneity both in systems interacting with a short ranged screened Coulomb interaction, as well as in systems with a long range logarithmic interaction between the particles.

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In liquids and glassy systems there has been considerable interest in dynamical heterogeneities which occur when certain regions of the sample have a higher mobility than the rest of the sample. Particle motion in these systems often occurs by means of correlated motion of a group of particles along a string like structure. [1]. Most of the studies of heterogeneities in 2D and 3D have focused on systems that are supercooled near the glass transition in simulations [2] and experiments [3,4,5]. A recent study considered a relatively simple system of a 2D dense monodisperse colloidal assembly which shows an ordered to disordered transition as a function of density [4]. In this system the colloids form a defect free triangular lattice at high densities and disorder for lower densities. Near the disordering transition at the close packing density, collective heterogeneous motion of the particles appears, and the system consists of domains of sixfold coordinated particles, $n_c = 6$, surrounded by grain boundaries composed of strings of $n_c = 7$ and $n_c = 5$ dislocations. The formation of grain boundaries or defect condensation in 2D monodisperse liquids has also been observed in colloidal assemblies [5,6] and in dusty plasmas [7,8,9].

Since the dislocations have a complex interaction with one another, consisting of a long range logarithmic repulsion and a short range attractive core interaction, one could expect complicated dislocation dynamics for systems with even a low density of dislocations. In the liquid state, creation and annihilation of dislocation pairs also occurs due to the thermally induced motions of the underlying particles. Although there have been numerous studies of the fluctuations of the particles in 2D liquids, the fluctuations in the *defect density*, defined as the number of particles with $n_c \neq 6$, have not been investigated. This measure would be easy to access in experiments on systems such as colloids and dusty plasmas where the individual particles can be directly imaged. It is not known how the density of $n_c \neq 6$ particles would fluctuate as the ordering transition is approached. If the defects are concentrated in clumps or grain boundaries, then it is likely that the creation or annihilation of defects will be highly correlated, which can give rise to $1/f^\alpha$ fluctuation spectra. Conversely, if the defects are appearing and disappearing in an uncorrelated manner, a white noise spectra would arise. We also wish to connect the formation of strings of defects with the appearance of correlated particle motion along 1D strings.

In this work we show that for 2D systems which form triangular lattices at low temperatures, for increasing temperature there is a transition from a non-defected regime to a defected regime where there is a proliferation of defects. Here we do not attempt to examine the nature of the disordering transition, such as whether there is an intermediate hexatic phase [10]. Instead, we concentrate on the motions of the particles and the defect fluctuations in the liquid phase. Close to the disordering transition, in the defected regime, the system consists of regions of particles with sixfold order surrounded by strings or grain boundaries of 5-7 fold defects, and there are few free dislocations. We also find that in this region, collective particle motion occurs in 1D strings along the grain boundaries. When temperature is fixed, the density and structure of the defect strings fluctuates with time. Near the disordering transition, the defect density shows large fluctuations with $1/f$ spectra. For increasing temperatures, the defect density increases and the string like structures of the defects and the 1D string like motion of the particles are lost. At the same time, the low frequency noise power increases and the spectrum crosses over to a white noise signature, indicating the loss of correlations in the creation or annihilation of defects.

We numerically study 2D systems at finite temperature using Langevin dynamics. We consider monodisperse particles in a sample with periodic boundary conditions and have investigated two types of interactions. Most of the results presented here are for particles interacting via a Yukawa or screened Coulomb interaction potential, $V(r_{ij}) = (Q^2/|\mathbf{r}_i - \mathbf{r}_j|)\exp(-\kappa|\mathbf{r}_i - \mathbf{r}_j|)$. Here $\mathbf{r}_{i(j)}$ is the position of particle $i(j)$, Q is the charge of the particles, $1/\kappa$ is the screening length, and we use

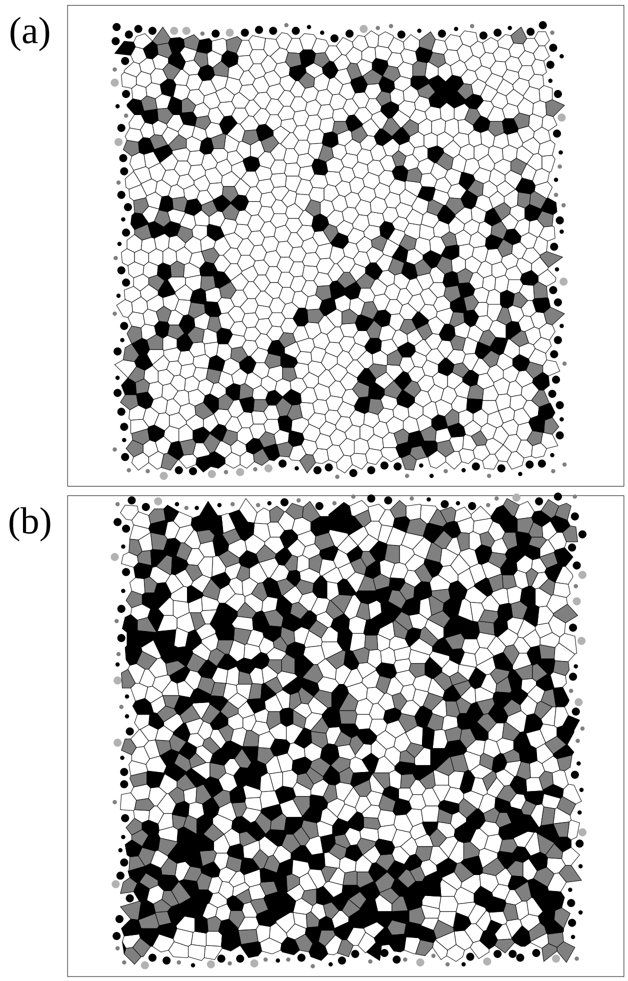

FIG. 1. The Voronoi constructions for a 2D system of particles interacting with a screened Coulomb potential. A particle is centered at each polygon and the polygon color is: $n_c = 6$, white; $n_c = 5$, dark gray, $n_c = 7$, black. (a) $T/T_d = 1.04$; (b) $T/T_d = 7.0$.

$\kappa = 2/a_0$, where a_0 is the lattice constant. We choose to study this interaction since the quantities we study in this work can be accessed in systems such as 2D colloidal assemblies and dusty plasmas, in which the particles interact via a screened Coulomb interaction. We also consider particles interacting via a logarithmic potential, $V(r) = \ln(r)$. Unlike the Yukawa potential, this interaction is long-range so that an interaction cutoff cannot be implemented. For increased computational efficiency we use the summation method of Ref. [11]. A physical example of logarithmically interacting particles with overdamped dynamics is vortices in thin-film type-II superconductors.

The equation of motion for particle i is:

$$\frac{d\mathbf{r}_i}{dt} = -\sum_{j\neq i}^{N_c} \nabla_i V(r_{ij}) + \mathbf{f}_T \tag{1}$$

where $\mathbf{f}_T$ is a randomly fluctuating force due to thermal kicks with $< \mathbf{f}^T(t) >= 0$ and $< \mathbf{f}^T(t)\mathbf{f}^T(t') >= 2k_BT\delta(t-t')$. In all the simulations we initially start from an ordered triangular configuration. We fix the temperature $\mathbf{f}_T$ for 10^6 time steps before we begin to take data. We note that previous simulations have shown that extremely long time transients can arise near the order to disorder transition of up to 2×10^6 time steps for system sizes of 36864 particles [12]. In this work we limit ourselves to system sizes of $N_c = 2000$ or less, so that 10^6 time steps is adequate for equilibration. In addition, histograms of the time series of the defect density are Gaussian, which is further evidence that our systems are equilibrated. We measure temperature in units of T/T_d where T_d is the temperature at which the first free dislocations appear.

In Fig. 1(a) we show the Voronoi construction of a system of particles with a screened Coulomb interaction for $T/T_d = 1.04$. The Voronoi construction is similar to the Wigner-Seitz cell construction and an individual particle is located in the center of each cell. If a particle has six nearest neighbors, $n_c = 6$, then the polygon has six sides. In Fig. 1 particles with $n_c = 6$ are white, $n_c = 5$ are dark gray, and $n_c = 5$ are black. We analyze a series of such images at fixed $T/T_d = 1.04$. We find that 29% of the particles are defected and that most (94.5%) of the defects are part of a cluster or condensate rather than free. In some regions, the defects form strings or grain boundaries. There are also a small number of free disclinations present, indicating that we are in the liquid phase rather than a hexatic phase, according to 2D continuous melting theories. Clustering of defects has been observed in experiments on dusty plasmas [7] which find comparable numbers of free dislocations. In Fig. 1(b) we show the same system at a higher temperature, $T/T_d = 9$. Here, the number of defects is higher with most of the defects again predominately appearing in clusters; however, there is no string like characteristic to the clusters. Instead, the defects form clump like objects.

In Fig. 2(a) we show the positions of the particles (black dots) and trajectories (lines) for the same system in Fig. 1(a). The trajectories are taken for the particle motion over 10000 time steps. Fig. 2(a) shows that the motion is heterogeneous with certain regions in which the particles do not move. These areas are also the defect free regions. In the areas where there is motion, the particles move by a, and there is some tendency for the motion to occur along string like paths. We also often find places where the motion occurs in a circular path with an immobile particle in the center. A similar kind of motion in 1D like strings and rotations was seen in experiments on dense colloidal liquids [4]. If we take trajectories for longer times, eventually all the particles take part in some motion. In Fig. 2(b) we show the positions and the trajectories for the high temperature phase in system Fig. 1(b) for 2000 time steps. Here the motion occurs everywhere with no evidence for correlated stationary regions. We find that even for very short times,

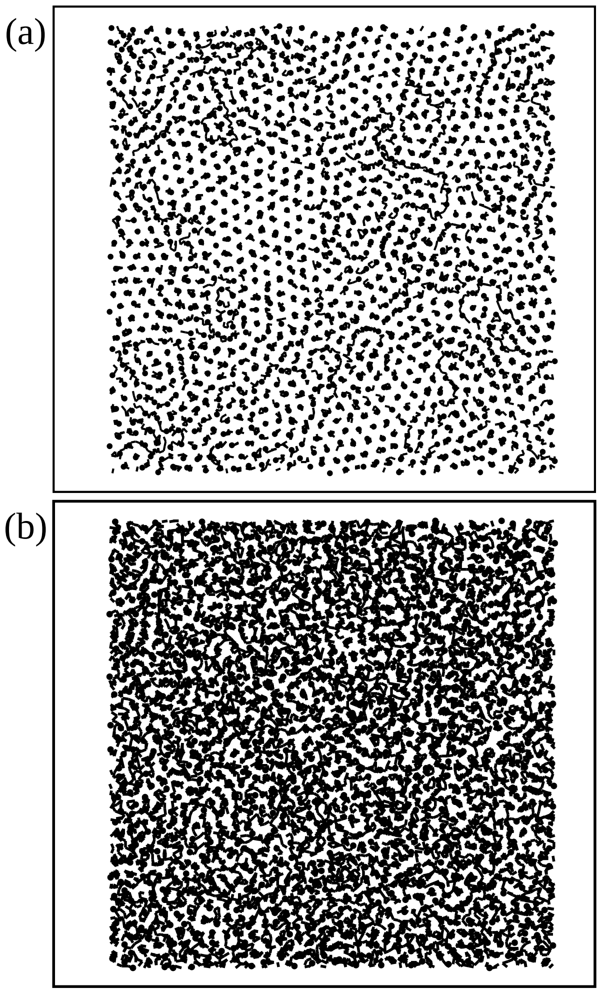

FIG. 2. Particle positions (black dots) and trajectories (black lines) for a fixed period of time for (a) $T/T_d = 1.04$ and (b) $T/T_d = 7.0$.

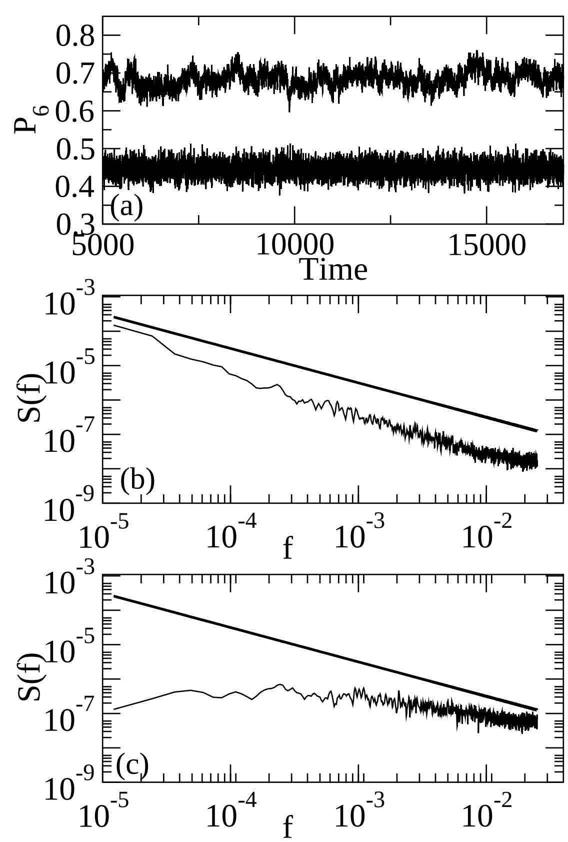

FIG. 3. (a) A portion of the time series of the fraction of $n_c = 6$ particles, $P_6(t)$, for (upper curve) $T/T_d = 1.04$ and (lower curve) $T/T_d = 7.0$. (b) The power spectrum for the time series at $T/T_d = 1.04$. The solid line indicates a slope of $1/f$. (c) The power spectrum for the time series at $T/T_d = 7.0$ along with a $1/f$ line.

there are no correlations in the motion.

We next consider a way to characterize the behaviors of the system at the different temperatures by measuring the fluctuations of the defect density as a function of time. We compute the defect configuration and density every 20 time steps for 20000 frames and obtain a time series of the defect density vs time for several temperatures from $T/T_d = 1.04$ to 10. We have considered a variety of sampling rates and find consistent results. In Fig. 3(a) we show a portion of the time series of the fraction of $n_c = 6$ particles, $P_6(t)$, for $T/T_d = 1.04$ (upper curve) and $T/T_d = 7.0$ (lower curve). For $T/T_d = 1.04$ the fluctuations show long time variations, while for the higher temperature the fluctuations are very rapid. In Fig. 3(b) we plot the power spectrum $S(f)$ of $P_6(t)$ for $T/T_d = 1.04$, which fits well to a $1/f^\alpha$ scaling over more than three decades with the best fit $\alpha = 1.04$, close to $1/f$ noise. As the temperature increases, the spectrum changes from $1/f$ to white noise ($\alpha = 0$) for low frequencies. The small frequencies correspond to the long time correlations in the system indicating that correlation times of the defect creation or annihilation decrease at higher temperatures. In Fig. 3(c), the power spectrum for $T/T_d = 7.0$ is white with $\alpha = 0$ for a large portion of the curve. We have also considered $P_6(t)$ for $T/T_d < 1.0$ where there are a small number of bound dislocation pairs present. Here we find white noise with small amplitude fluctuations.

The magnitude of the noise power, obtained by integrating $S(f)$ over the first octave of frequencies, is related to the defect density and the disordering transition. In Fig. 4(a) we show P_6 as a function of T and in Fig. 4(b) we show the corresponding noise power S_0. The peak in S_0 coincides with the onset of the defect proliferation. For increasing T, the noise power falls and saturates when the spectrum becomes white. There is a finite S_0 for $T/T_d < 1.0$ since, as noted, pairs of bound dislocations can still be thermally created. We have repeated the same set of simulations for particles interacting with a long range logarithmic potential and again find a noise power peak coinciding with the defect proliferation

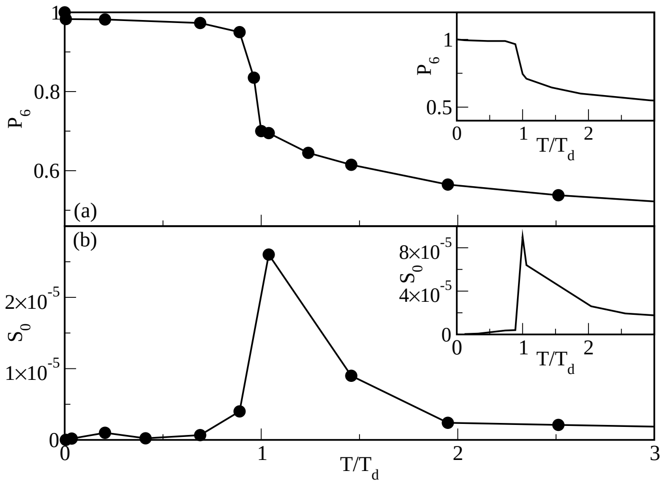

FIG. 4. (a) The fraction of six-fold particles P_6 vs T for a system of particles with a screened Coulomb interaction. Inset: P_6 vs T for a system with logarithmic interactions. (b) The integrated noise power S_0, obtained from $S(f)$, vs T for the same system in (a). Inset: S_0 vs T for a system with logarithmic interactions.

(insets of Fig. 4) and a $1/f$ power spectrum just above T_d.

We have also considered a sudden quench from $T/T_d > 1.0$ to $T/T_d < 1.0$. Here the system is out of equilibrium, and approaches equilibrium by means of the annihilation of the defects. In Fig. 5 we show the particle motions after a quench from $T/T_d = 1.04$ to $T/T_d = 0.2$. We find that the number of defects decreases rapidly to a saturation point where a few defects or grain boundaries remain. The motion of the particles corresponds to the annihilation of the defects. As seen in Fig. 5, the motion during the defect annihilation process has the same string like nature as the equilibrium $T > T_d$ motion near the defect proliferation transition. This result suggests that the heterogeneous particle motion is produced by the motion of defects, particularly the creation or annihilation of these defects in a correlated manner.

In summary, we have proposed a new way to examine dynamical heterogeneities in a liquid by measuring the fluctuations in the topological defect density. We find that near the onset of defect proliferation, the particle motion is heterogeneous and the defects cluster together into grain boundaries or strings. The defect fluctuations in this phase have a $1/f$ character and a large noise power. For higher temperatures the heterogeneities are lost and the fluctuation spectrum is white. We have also examined the defect annihilation after a quench from the disordered phase to the ordered phase and find that the particle motion also shows heterogeneities as the defects are annihilated. We have considered systems with screened Coulomb interactions and logarithmic interactions and find similar behavior. Our predictions can be easily tested in 2D dense colloidal liquids. It would also be interesting to study the fluctuations of defects in 3D. This would be experimentally possible in 3D colloidal assemblies using confocal microscopy.

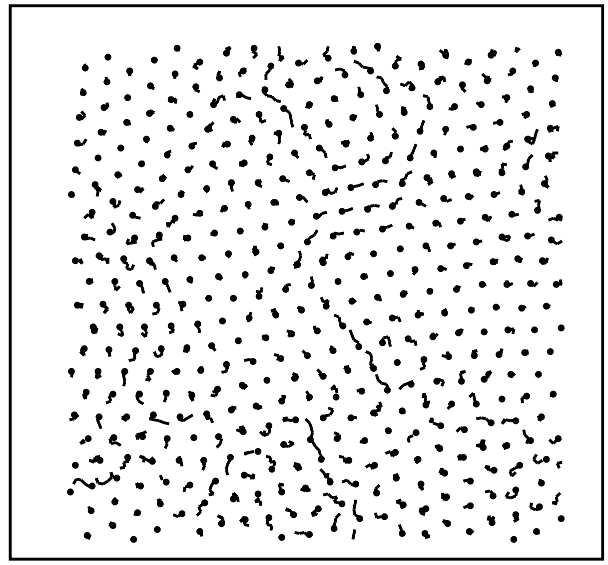

FIG. 5. The particle positions (black dots) and trajectories (black lines) for a system quenched from $T/T_d = 1.04$ to $T/T_d = 0.3$. The trajectories are analyzed once the system is at the lower temperature. During this time a portion of the defects are annihilated.

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